ARKIV FÖR FYSIK Band 19 nr 22

Communicated 7 December by The Svedberg and Erik Rudberg

Nuclear spectroscopy of mass-separated neutron-deficient tellurium isotopes

By R. W. Fink,[1] G. Andersson, and J. Kantele[1]

With 14 figures in the text

ABSTRACT

The radioactive decay of five neutron-deficient activities of tellurium from irradiations of Sb with 90–140 MeV protons has been investigated with magnetic and scintillation spectrometers, beta proportional counter, and alpha ionization chamber using mass-separated sources.

The electron spectra and the gamma scintillation singles and coincidence spectra have been interpreted in the form of tentative disintegration schemes to account for the decay modes of Te^{116} (2.50 ± 0.02 hours), Te^{117} (61 ± 2 minutes, measured spin 1/2), Te^{118} (6.00 ± 0.02 days), and Te^{119} isomers (15.9 ± 0.3 hours, measured spin 1/2, and 4.5 ± 0.3 days, measured spin 11/2). Two neutron-deficient Sb isotopes also were prepared by mass separation and their half-lives confirmed: Sb^{115} (36 ± 3 minutes) and Sb^{116} (64 ± 4 minutes). From NH_4I targets irradiated with 145–180 MeV protons, a new Te activity was found with half-life about 5–6 minutes, which might be Te^{115} or Te^{114}. A new carrier-free radiochemical separation technique requiring about 30 seconds is reported for preparing Te sources from the irradiated NH_4I targets.

No isomer in Te^{117} having a half-life greater than 15 minutes was detected, and in the case of the Te^{119} isomers, no transitions corresponding to the internal branch could be found, the two isomers apparently decaying independently to levels in antimony.

1. Introduction

Some years ago the neutron-deficient isotopes of tellurium were surveyed briefly in connection with spallation studies. Following the discovery of 4.5 day Te^{119} from the $Sb(d, 4n)$ reaction at 40 MeV by Lindner and Perlman [1], who showed it to be the parent of 38 hour Sb^{119}, an isomeric state of 16 ± 2 hours half-life was found by Dropesky [2] in 240 MeV proton reactions on iodine when the tellurium fraction was followed with an integral-bias-type scintillation counter, and also as a daughter of 18 min I^{119}. This activity also was observed in proton spallation studies of cesium [3]. The 6.0 day Te^{118} was found by Lindner and Perlman [1] to be the parent of 3.5 min Sb^{118}. Conversion electrons below about 1 MeV could not be detected in a crude magnetic spectrometer. Low-energy positrons of about 0.7 MeV end-point in coincidence with a 108 keV gamma were thought to have been observed [3] by beta–gamma coincidence absorption experiments, but this result was inconclusive and probably wrong owing to the presence of Te^{119} and other species in the sample. A strong 2.5 hour activity also was found [1, 2, 3] but was incorrectly assigned to Te^{117}. In the present work it is shown by mass separation to be Te^{116}. An activity of the order of 3 hours, the parent of a 10–20 min Sb activity, was assigned to Te^{116} by Dropesky [2]. More recently the half-life of Te^{116} was determined by successive radiochemical separations of the antimony daughter and a value of 2.5 hours found [4]; in similar experiments, a value of about 1.7 hours was reported for Te^{117} [4]. In bombardments of indium with 48 MeV alpha particles and of tin with 24 MeV deuterons and 12 MeV protons, some unassigned activities were found by Rhodes [5] in the radiochemically separated tellurium fraction with half-lives of 16 min, 1.4 hours, and 7.5 hours, and in the antimony fraction with half-lives of 7, 30, 60 min, and 4 hours. New antimony isotopes Sb^{113}, with half-life 7.0 ± 0.5 min (2 MeV β^+) from the $Sn^{112}(d, n)$ reaction at 10 MeV on 52.3 % enriched Sn^{112}, and Sb^{115}, with half-life 31 ± 1 min (2 MeV β^+, no gammas > 0.5 MeV) from the $Sn^{114}(d, n)$ reaction at 10 MeV on 57.2 % enriched Sn^{114}, were found after radiochemical separation [6]. The decay schemes of 60 min Sb^{116} and 5.1 hour Sb^{118} isomers have been investigated in some detail at Copenhagen [7] for comparison with the theoretical calculations by Kisslinger and Sorensen [8] of the effect of a pairing and long-range force interaction on the structure of two-particle excitation in even–even nuclei with a single closed shell, e.g. tin. Glendenning [9] has discussed recently some theoretical predictions concerning levels in odd-mass tellurium and xenon nuclei. During the course of the present investigation, the decay scheme of the Te^{119} isomers has been studied with magnetic and scintillation spectrometry on non-mass-separated samples [10].

---

[1] Present address: Department of Chemistry, University of Arkansas, Fayetteville, Arkansas, U.S.A.

The present study was initiated because the availability at the Werner Institute in Uppsala of a mass separator, 190 MeV synchrocyclotron, and high-resolution beta spectrometer and at the Institute of Physics in Helsinki of scintillation coincidence spectrometers with 5 × 4 inch NaI(Tl) detectors and a multichannel analyzer[1] afforded an opportunity for a penetrating investigation of this region with the possibility of extending knowledge of the system of isomers in odd-A tellurium nuclei.

2. Experimental

2.1. *Preparation of sources*

Irradiations of 2 to 6 hours with 90 to 140 MeV protons on 400 mg "specpure" antimony metal powder were carried out. The peak yield among the tellurium isotopes occurred at Te^{118} when the bombarding energy was 100 MeV, and at Te^{119} with 90 MeV. The target was dissolved in 1 ml concentrated HF containing from 2 to 12 mg Te(VI) carrier by adding concentrated HNO_3 dropwise until the antimony dissolved completely. This solution was evaporated to dryness to expel HF and HNO_3, followed by evaporation twice to dryness with concentrated HCl, reducing tellurium to Te(IV). The residue was taken up with heat in 3 ml $3N$ HCl and diluted to about $2N$. A few milligrams of hydrazine hydrochloride, $NH_2NH_2 \cdot HCl$, were added, and SO_2 gas was bubbled through the solution, which was heated in a boiling water bath, in order to precipitate tellurium as the metal, a drop of Kodak "Filterflo" wetting agent (dilution 1:100) solution being added to improve the coagulation properties of the Te precipitate. After centrifugation and decantation of the supernatant liquid containing Sb and Sn activities, etc., the Te precipitate was washed twice with 3N HCl (containing a drop of wetting agent). The total time required to bring the separation to this point is about 30 min. The decontamination from other activities is in the region of 10^3 to 10^4.

---

[1] We are indebted to Prof. L. Simons and his group at the University of Helsinki for graciously permitting us the use of this scintillation equipment.

For the mass-separator ion source [11] the Te metal is converted in about 10 minutes' additional time to $TeBr_4$ by dissolving the metal in 0.5 ml aqua regia, evaporating twice to dryness with concentrated HCl, then taking up in concentrated HBr and again evaporating to dryness to obtain reddish-orange $TeBr_4$ (10 to 12 mg is usually the best quantity for the ion source used). The $TeBr_4$ is dissolved in ethanol for transfer to the ion source crucible, which is warmed under an infra-red lamp. $TeBr_4$ has the desirable property, in addition to its complete solubility in ethanol, of subliming at low temperature (300–400°C), giving a suitable vapor pressure under the normal ion source conditions and smooth operation with little or no sparking. In this respect it is far superior to Te metal as a charge material. The astonishingly high value of about 24 % was obtained in a yield experiment for the mass-separator efficiency.

In the tellurium region, the spacing between adjacent masses in the Werner Institute separator is of the order of 6 mm. The purity of the separated sources is discussed individually below. The general properties of such mass-separated sources for use with the beta spectrometer of the Institute have been discussed recently [12].

For experiments with non-mass-separated Te sources, two other radiochemical techniques were employed, one being electrodeposition of carrier-free Te requiring about 90 min, and the other a new method to be described below requiring about 30 sec. In the former method, carrier-free Te was electrodeposited [13] onto 5 mg/cm^2 copper foils for use as beta spectrometer sources. Difficulties were encountered, however, since a black, spongy deposit forms on the cathode when a solution of hydrazine hydrochloride or HCl is electrolyzed between a platinum anode wire and copper or nickel cathode foils. Attempts to eliminate the black deposit were successful only when platinum foil was used as the cathode. However, of the order of 25 % or less of the carrier-free Te(IV) could be electrodeposited in about 10 min from a solution containing about 1 mg hydrazine in dilute HCl added to increase the conductivity to about 1 A at 15 V, but if the electrolysis was continued longer, serious losses occurred owing to the flaking off of the black deposit (in one such experiment the amount of Te activity deposited amounted to only 4.5 %). It is probable that the troublesome black deposit consists of finely powdered metal which is first dissolved from the cathode surface by action of hydrogen ions and then is reduced to metal by nascent hydrogen atoms. This would explain the absence of the black deposit on platinum cathodes which are not attacked by hydrogen ions.

Methods for the preparation of carrier-free Te by anion exchange are reported [14]. In the present investigation, however, carrier-free Te was prepared from antimony targets by carrying Te on 20 mg Se(VI) carrier added to the concentrated HF–HNO_3 solution used to dissolve the target. The Se was precipitated as the element from 2N HCl solution, similar to the tellurium procedure given above, with SO_2 gas and hydrazine solution in a boiling water bath. The red Se° precipitate tends to be colloidal and difficult to centrifuge down unless it has been thoroughly coagulated by heat and wetting agent. After washing the precipitate twice with 3N HCl, it is dissolved in a few drops of aqua regia and evaporated to dryness twice with concentrated HCl. The precipitate is then taken up in concentrated HCl with heat and the solution cooled to 0°C in an ice bath. Gaseous SO_2 is then bubbled through until precipitation of Se° is complete. Carrier-free Te(IV) remains in the supernate, which is evaporated to dryness in order to expel the last traces of Se carrier as volatile $SeBr_4$. The carrier-free Te remaining is then taken up with heat in a few drops of concentrated HCl and evaporated to dryness to expel HBr. Finally it is taken up in distilled water or 0.1 *N* HCl as the final carrier-free aqueous Te fraction.

A new separation of carrier-free Te from proton irradiated NH_4I targets was devised in order to search for short-lived isotopes (e.g. Te^{115}, Te^{114}). With this technique it is possible to separate Te carrier-free from iodine, antimony, and tin activities in 30 sec or less. The NH_4I target is dissolved in a few drops of hot water containing a few milligrams of hydrazine hydrochloride, and the solution is quickly evaporated to dryness on a clean platinum surface and ignited for about 1 sec to a red heat in the flame of a Fisher burner. Interfering activities of iodine, antimony, tin, etc. are volatilized, leaving extremely clean, very intense, carrier-free Te sources. This fast procedure gives a high yield (at least 75 %) of the tellurium activity in the target.

In order to study neutron-deficient antimony isotopes, a fast separation of Sb from a tin target was employed, in which the tin target is dissolved in hot, concentrated HCl. From 1 to 10 mg Sb(III) carrier, as desired, is added and then reduced with metallic tin filings. A finely divided, black precipitate of Sb is obtained which is centrifuged off, washed with concentrated HCl, and mounted for use in the mass separator ion source. About 20 min are required. The metallic Sb, however, was not ideal for the ion source, but no better charge material could be found.

2.2. *The measurements*

Conversion electrons above 9 keV in energy were measured in a two-directional focusing beta spectrometer constructed by Arbman and Svartholm [15]. The spectra were recorded either by a linear rate-meter and Speedomax recorder in scanning runs or by automatic step-by-step registration equipment [16]. Two different GM detector windows were used, one of approximately 50 $\mu g/cm^2$ VYNS plastic [17] for energies down to 9 keV, and the other of 0.9 mg/cm^2 Mylar for energies above about 50 keV, although the thinner window was used in the high-energy region as well. The resolutions generally used in conversion electron measurements were in the range of 0.35 to 0.5 %. Positron spectra were taken with the transmission set to about 2 %, corresponding to about 1 % resolution.

Gamma singles and coincidence spectra were measured with two Harshaw NaI(Tl) crystals, 5 inches in diameter by 4 inches deep, mounted on DuMont 6363 photomultiplier tubes, 3 inches in diameter, connected to linear non-overloading amplifiers of Hamner and Franklin manufacture. The pulses were analyzed by a 100-channel Hutchinson–Scarrot kick-sorter built by Marshall Electric Co., England. The coincidence analyzer was type 1036 (Dynatron Radio Ltd., England) with variable delay and resolving times. The crystals were shielded by 5 to 10 cm of lead. The source-to-crystal distance generally was 20 cm, although this distance was suitably modified in order to accommodate stronger and weaker sources. The resolution was 8.6 % for the 661 keV Cs^{137} photopeak and 5.6 % for the 2.62 MeV photopeak from ThC″. Counts falling in the lowest channels (below channel 15) were corrected, according to a method described previously [18], for "damping", a type of reproducible malfunction of the analyzer. Background was subtracted automatically from all spectra, but in certain cases when the background was strong compared with the spectrum, it could not always be strictly subtracted. When the total counting rate in the detector rose above a critical limit, it affected the position of the photopeaks, shifting them toward higher energy with increasing counting rate. Great care was taken, therefore, to keep the total counting rate below this limit, and for precise energy determinations to make the measurements at various counting rates encompassing the counting rate of the standard sources used for energy comparison. The linearity of the spectrometer was excellent in the region needed, and the stability was good at night when the data were taken. The spectra were calibrated against standard sources of Te^{123m} (159.2 keV), Hg^{203} (279.1 keV), Cs^{137} (661 keV), Na^{22} (511 and 1273.6 keV), Mn^{54} (835 keV), Co^{60} (1172.6 and 1332.9 keV), and ThC″ (2614 keV). Rapid, repeated comparisons were made alternately between sample and standard, care being taken to keep the total counting rates equivalent and well below the critical limit. A single-channel analyzer and scaler also were used to check some energy values. For coincidence measurements the second detector was positioned axially opposite the first one, the geometry used being indicated in the figures representing specific coincidence spectra. A simple gate generator modified pulses from the coincidence unit for gating the analyzer. The resolving time generally used was 0.5 μsec, the accidental coincidences being subtracted with the 1 μsec delay "reversed". Although the background always was negligible, in many cases the spectrum was so complicated that it was impossible to obtain a "pure" spectrum coincident with a given gamma line. In a few cases the influence of extraneous gammas could be corrected for by shifting the lower limit of the window by an amount equal to the width of the window, the spectrum thus obtained then being subtracted from the primary spectrum. This accordingly makes the relative intensities obtained from coincidence spectra somewhat uncertain.

All gamma measurements were performed at least five times using mass-separated sources from six bombardments, except in the case of 61 min Te^{117} whose coincidence spectrum could be obtained in only one run. Results given below are the weighted means from the best runs.

Positron counting was done by means of annihilation quanta, the scintillation detector being calibrated at identical geometry with Na^{22}, which exhibited efficiency-corrected relative intensities for the 511 and 1274 keV peaks of 443:172, respectively; correcting this ratio for 10 % electron capture, this becomes 443:155. At the geometry used therefore, each Na^{22} positron corresponds to 2.86 annihilation quanta. (This number decreased about 5 % when the source-to-crystal distance was increased by an additional 20 cm.)

Gamma spectra were resolved by using response functions (standard gamma shapes) from the calibration sources, relative intensities being obtained from the net corrected areas of the various photopeaks. Values calculated by Gove and Rutledge [19] were used to correct the intensities for detector efficiency. To check the reliability of the procedure, the ThC″ 2.62:0.58 MeV gamma relative intensities were measured with results in satisfactory agreement with the literature.

In determining the relative intensities of the gammas of about 2.1 MeV in Te^{116}, Te^{117}, and Te^{119} decays, the response function of the ThC″ 2.62 MeV gamma was used. Unfortunately, this shape is not accurate, because the pair production cross-section of NaI decreases by a factor of 2 as the gamma energy decreases from 2.6 to 2.1 MeV [20]. At the same time however, the total Compton cross-section increases, partly compensating this effect.

Gross beta decay was followed with a flow proportional counter fitted with an end-window of 250 μg/cm^2 gold-coated VYNS film [17]. A resolving time of about 9.3 μsec was measured for it by the paired source method. With a 10 % methane–90 % argon mixture and anode wire 0.002 inch in diameter, the operating voltage was 2250 V and the plateau 250 V in length.

A gridded ionization chamber filled to several atmospheres with 10 % methane–90 % argon was used to search for alpha emission from mass-separated tellurium sources, since conceivable alpha branching would lead to a closed shell ($Z = 50$) product. Very fast alpha pulses (0.1 μsec rise-time, delay line clipped to about 0.5 μsec duration) were obtained in order to avoid beta pile-up and were displayed on a Tektronix oscilloscope. The traces were photographed either with long exposures of single plates or else with a moving-film camera, by which every alpha pulse could be observed, so that the results could be analyzed both for energies and half-lives. These alpha detection experiments will be published in detail separately [21]. Limits for alpha branching were set, representing the maximum sensitivity of the chamber for alpha particles above 1.9 MeV in the presence of intense beta–gamma radiation, which in fact was the limiting factor on the ultimate sensitivity owing to insulation "breakdown" which appeared when sufficiently intense beta–gamma radiation was present.

3. Results

3.1. Te^{116}

Following a clearly evident initial growth, due to 15.5 min Sb^{116} daughter, gross beta counting of mass-separated Te^{116} sources in the beta proportional counter exhibited very straight decay over some 5 half-lives before tailing out to weak long-lived contaminants and background. Typically the amount of (4.5 + 6.0 day) Te^{119} and Te^{118} contamination in these sources was less than 0.5 % of the initial Te^{116} activity, whose half-life was determined to be 2.50 ± 0.02 hours.

A representative electron spectrum is shown in Fig. 1, taken in the beta spectrometer. Lines can be seen which arise from K Auger and two gamma transitions (see Table 1) at 93.9 ± 0.2 and 101 ± 1 keV. Table 1 summarizes the energy data concerning these lines. The electrons in the 21.0 to 26.0 keV region, which follow a 2.5 hour decay, are interpreted to be the K Auger lines. A similar assignment of lines in this energy region has been made below in the case of Te^{119}, in accordance with the recent calculations of Asaad and Burhop [22] and experimental values of Johnson and Foster [23] at $Z = 47$ and Marguin and Moussa [24] at $Z = 56$. The principal K Auger transitions and expected relative intensities are listed in Table 3, with which a comparison of Fig. 1 (left) supports this assignment.

In the case of the 93.9 keV transition, the experimental ratio $K/\Sigma L = 2.88 \pm 0.16$, which agrees with $E2$ multipolarity by comparison with Rose's theoretical value of 2.88 [25].

No trace of lines from the 137 or 404 keV transitions from 60 min Sb^{117m} decay was detected, and it is clear that this high-spin isomer is not fed detectably in the decay of Te^{116}.

The relative intensities of the two transitions lie in the ratio of about 76:1 for the 93.9 and 101 keV transitions, respectively, on the assumption of either $M1$ or $E2$ multipolarity for the latter.

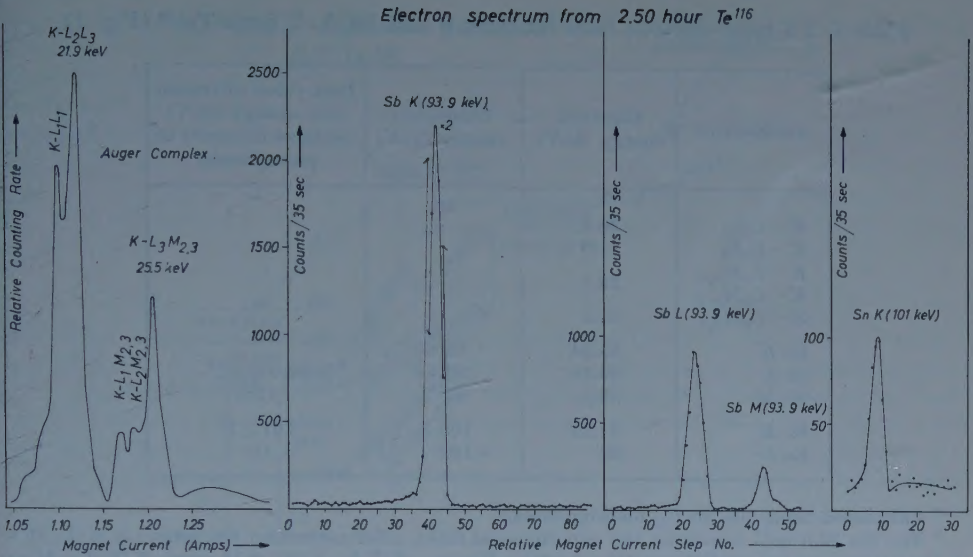

Fig. 1. Composite plot of representative electron spectra from 2.50 hour mass-separated Te^{116} taken in the two-directional focusing beta spectrometer. A thin window ($\sim 50\ \mu g/cm^2$ VYNS) GM detector was employed in these runs. The K Auger spectrum (left) was traced from an automatic run with rate-meter–Speedomax recording at approximately 0.5 % resolution. The energies and relative intensities fit an assignment of these lines better as the K Auger spectrum than as a possible 25.8 KeV gamma transition in antimony. The K, L, and M lines arising from a 93.9 keV ($E2$) transition in antimony were taken at higher resolution (approximately 0.35 %) using step-scanning and decimal print-out scaling equipment. Similarly, the K line from a 101 keV transition, tentatively assigned in tin, was measured. The L line from the 101 keV transition is not shown here but was measured in other runs. In this plot the relative intensities of the K, L, and M lines of the 93.9 keV and the K line of the 101 keV transition ($\times 10$) are directly comparable.

The strong positron spectrum, which decayed with a half-life of about 2.5 hours, appeared to be complex with an end-point of 2.27 ± 0.10 MeV, due to the 15.5 min Sb^{116} daughter, and a possible low-energy group, conceivably due to Te^{116}, having an end-point of about 0.44 MeV in an allowed FK plot, although this is rendered somewhat inconclusive by the question of scattering and by poor statistics.

For the 2.27 MeV positron transition to the 1292 keV level in tin, $\log ft = 5.1$, and for the beta transition to the 2230 keV level, $\log ft = 4.3$ (cf. Fig. 2). Thus, both of these transitions, as well as the 0.44 MeV positron transition from Te^{116} ($\log ft \sim 4.7$), are allowed. From the relative gamma intensities given below, the total $(EC + \beta^+)$ branching from 15.5 min Sb^{116} may be computed to be 41 % to the 2230 keV level in tin and 59 % to the 1292 keV level. A total (EC/β^+) ratio of about 3.5 for these two transitions taken together is calculated, but this disagrees with the theoretical K/β^+ ratios for allowed transitions of 2.3 and 0.45, respectively [26]. Since these transitions clearly are allowed from the log ft values, the reasons for the disagreement in the EC/β^+ ratios are not understood, since L-capture cannot contribute more than about 15 % to the theoretical ratios [27].

A careful search for conceivable alpha emission from very strong mass-separated sources in the alpha ionization chamber enabled an upper limit to be set against alpha emission of $\alpha/\beta < 10^{-9}$ for Te^{116}.

Table 1. 2.5 hour electron lines between 9 and 350 keV from Te^{116} (Fig. 1).

Assignment	Electron energy (keV)	Transition energy (keV)	Best value of transition energy (keV) (relative intensity in parenthesis)
$K - L_1L_1$	20.9		
$K - L_2L_3$	21.9		
$K - L_1M_{2,3}$ $K - L_2M_{2,3}$	23.8		
$K - L_3M_{2,3}$	25.5		
Sb K	63.34	93.84	
Sb L	89.55	93.94[a]	93.9 ± 0.2[b]
Sb M	93.5	94	(76)
Sn K	71.12	100.3	101 ± 1[c]
Sn L	98	~102	(1)

[a] Calculated using the Sb L_{II} binding energy.

[b] For the 93.9 keV transition the experimental ratio, after correction for decay, is $K:L:M = (0.405 \pm 0.010):(0.141 \pm 0.007):(0.036 \pm 0.004)$. Hence, $K/\Sigma L = 2.88 \pm 0.16$ and $K/\Sigma(L+M) = 2.28 \pm 0.15$ in agreement with an $E2$ transition in Sb.

[c] Assignment to Sn uncertain; see discussion in text.

The gamma singles spectra measured with a 5 × 4 inch NaI(Tl) crystal, exhibited the following gammas and relative intensities (uncorrected for conversion and given in parentheses): 511 ($\gamma^{\pm}$) (123), 930 ± 10 (40), 1292 ± 8 (130), and 2230 ± 25 (22) keV, the relative intensities being good to within 20 %. Since 2.86 annihilation quanta per positron were detected under the present geometry (see 2.2), the total positron relative intensity thus is 43.

There is a disagreement in the case of the 1292 keV transition with an energy value of 1274 ± 6 keV obtained by Slätis, DuToit, and Siegbahn [28] from the decay of In^{116m}. Consequently, a careful intercomparison with the Na^{22} standard was carried out on two Te^{116} sources, one of which exhibited a total crystal counting rate slightly higher than that of the standard and the other, lower. In both cases the energy agreed within 2 keV and lay clearly higher than the Na^{22} energy which was taken as 1273.6 ± 1.6 keV from a recent measurement [29]. Also a further measurement was made on another Te^{116} source from a different bombardment by using Co^{60} as the comparative standard. This agreed precisely with the first result. All counting rates were kept well below the level where shifting of the photopeaks toward higher energy with increasing counting rate occurred. Stähelin, Maeder, and Pochon [30] have measured this transition from 15.5 min Sb^{116}, obtaining a value, corrected for the new Na^{22} energy, of 1302 ± 15 keV in agreement with the present result. From the decay of 54 min In^{116m}, Girgis and Van Lieshout also measured this transition energy, obtaining a value of 1300 ± 10 keV [31].

The branching ratio 930 keV gamma/2230 keV gamma = 64/36, the intensity of the 2230 keV gamma being more than twice that reported by Stähelin *et al.* [30] from a small crystal study of 15.5 min Sb^{116}. However, the efficiency calibration of the large (5 × 4 inch) crystal was confirmed by measuring the relative intensity ratio of the 0.58 MeV and 2.62 MeV gammas from ThC″ which agreed with the literature.

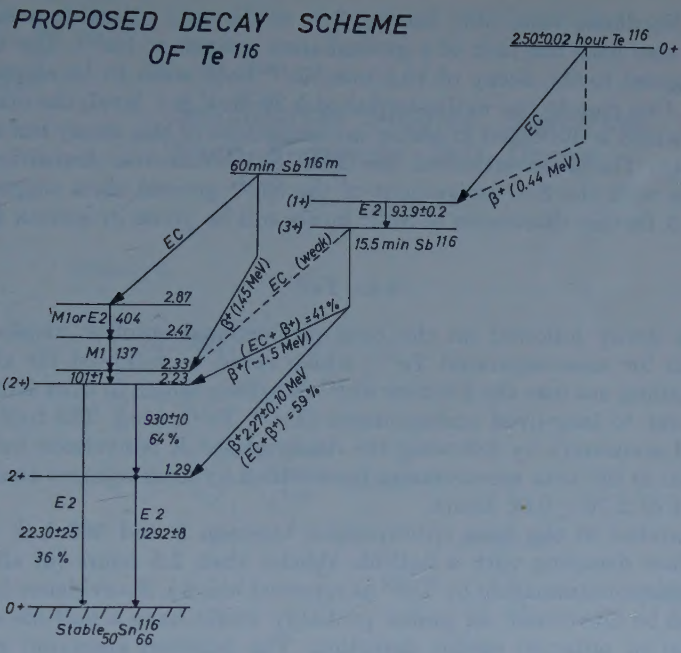

Fig. 2. Decay scheme proposed for Te^{116}. A spin assignment of 3 + for 15.5 min Sb^{116} appears to agree well with the intensities of transitions leading to and from this state. The 60 min Sb^{116} state is not fed to a detectable degree in Te^{116} decay, as shown by the absence of the 137 and 404 keV transitions in either the gamma scintillation or electron line spectra. The positron transitions exhibit allowed log *ft* values. The weak 101 ± 1 keV transition has been placed in tin, in agreement with previous work on the 60 min Sb^{116} decay, but it is also possible that it belongs in antimony. The portion of the Sn^{116} level scheme above the 2.23 MeV level is based on the one shown in Nuclear Data Sheets and represents a simplification of the one discussed in [7].

Gamma coincidence studies on Te^{116} with two 5×4 inch NaI(Tl) crystals confirm the results from studies of 15.5 min Sb^{116} sources [30]. A tentative decay scheme is proposed in Fig. 2, in which the 60 min Sb^{116} decay is drawn in even though it is not fed in Te^{116} decay (absence of 137 and 404 keV transitions). From the present results, the element assignment of the weak 101 keV transition is not clear. Since a transition of approximately this energy is already known in tin [7], we have identified it tentatively with the 101 keV transition, as shown in Fig. 2, although the conversion line energies seem to favour a higher Z for the latter (Table 1). It should be remarked that this part of the decay scheme is mainly based on the one in the Nuclear Data Sheets [32] and represents a simplification of the one discussed in [7].

The spin-parity assignments in Fig. 2 are based on the following arguments. Although the absence of a ground-state branch from 0 + Te^{116} is not proven, it seems reasonable to assume essentially 100 % branching to the 94 keV level in Sb^{116}. As discussed above, the 101 keV transition may belong in the Sb^{116} level scheme, but it is too weak to invalidate the assumption. With the positron energy indicated, log *ft* becomes allowed, establishing the 94 keV level as either 0 + or 1 +. Since it seems rather certain that the 94 keV transition is pure $E2$, the ground state of Sb^{116} would most likely be 2 + or 3 +. The former assignment, however, violates the Nordheim rules [33], leaving 3 + as the only plausible possibility. This is consistent also with the lack of a ground-state branch to Sn^{116}. The two positron branches assigned to the decay of 15.5 min Sb^{116} both seem to be allowed from the log ft values. One goes to the well-established 1.29 MeV 2 + level, the other to a level at 2.23 MeV which is indicated in earlier investigations of this decay but not assigned with certainty. The observation of the 2230 keV cross-over transition from this level together with the 3 + assignment of the Sb^{116} ground state suggests 2 + also in this case. A further discussion of Sn^{116} levels will be given in section 4.3.

3.2. Te^{117}

Gross beta decay followed on the beta proportional counter yielded a half-life of 61 ± 2 min for mass-separated Te^{117}, which could be followed for about 4 half-lives before tailing out into the 2.8 hour Sb^{117} daughter, which in turn after three half-lives tailed out to long-lived contaminants (Te^{118}, Te^{119}, etc.). The half-life of Sb^{117} was obtained accurately by following the decay of the K conversion line of the 161 keV transition in the beta spectrometer from which by least squares analysis a value was obtained of 2.76 ± 0.06 hours.

Careful searches in the beta spectrometer between 9 and 350 keV revealed no conversion lines decaying with a half-life shorter than 2.5 hours (in all cases there was appreciable contamination by Te^{116} as reported below). No evidence for isomerism in Te^{117} could be uncovered; an isomer probably would have a half-life shorter than about 15 min in order to escape detection. The positron spectrum exhibited an end-point of 1.74 ± 0.06 MeV ($\log ft = 5.1$) and an allowed FK plot revealed the possible existence of an inner group of end-point ~ 0.7 MeV, but this inner branch is rendered somewhat inconclusive by poor statistics. The positron spectrum decayed initially with a half-life of very roughly the order of an hour.

From alpha spectroscopy an upper limit of $\alpha/\beta < 5 \times 10^{-5}$ could be set against conceivable alpha decay of Te^{117}.

The gamma singles spectrum, Fig. 3, studied 6.8 hours after mass separation, exhibited the following gamma energies and relative intensities (uncorrected for conversion): 160 ± 5 (104 ± 25), 511 ($\gamma^{\pm}$) (17.8 ± 4), 718 ± 10 (9.1 ± 2), 1290 (1.3 ± 0.3), 1715 ± 20 (1.4 ± 0.2), 2240 ± 35 (1.0 ± 0.2) keV, and a complex between 0.9 and 1.1 MeV. The peak at 1715 keV is due in part to escape of one annihilation quantum from the crystal owing to pair production from the 2240 keV gamma, but the above relative intensity has been corrected for this effect. The 160 keV gamma decayed with a 2.8 hour half-life (Sb^{117}), and all of the others exhibited a 1 hour decay (Te^{117}). Comparison of this spectrum with that of Te^{116} indicated that the activity of the latter contaminating the Te^{117} source at the end of mass separation typically amounted to about 0.7 %, but at the time of measurement, this constituted a contribution from Te^{116} contamination of 45 % to the 1290 keV peak and of 15 % to the 2240 keV peak, making a precise energy determination of the former uncertain. In the "0.9–1.1 MeV" complex the sharpest part lies at 920–930 keV and appears to decay with a 1 hour half-life; consequently, a 930 keV transition is fitted into the decay scheme, which can accommodate such a transition (Fig. 4).

The coincidence spectrum was investigated with two 5 × 4 inch Nai(Tl) crystals. Owing to the short half-life, only qualitative data on the coincidence spectrum could be obtained. These are summarized in Table 2. The (0.51, 0.72) coincidence is predominant, indicating that most of the positron feeding goes to the 718 keV level. The

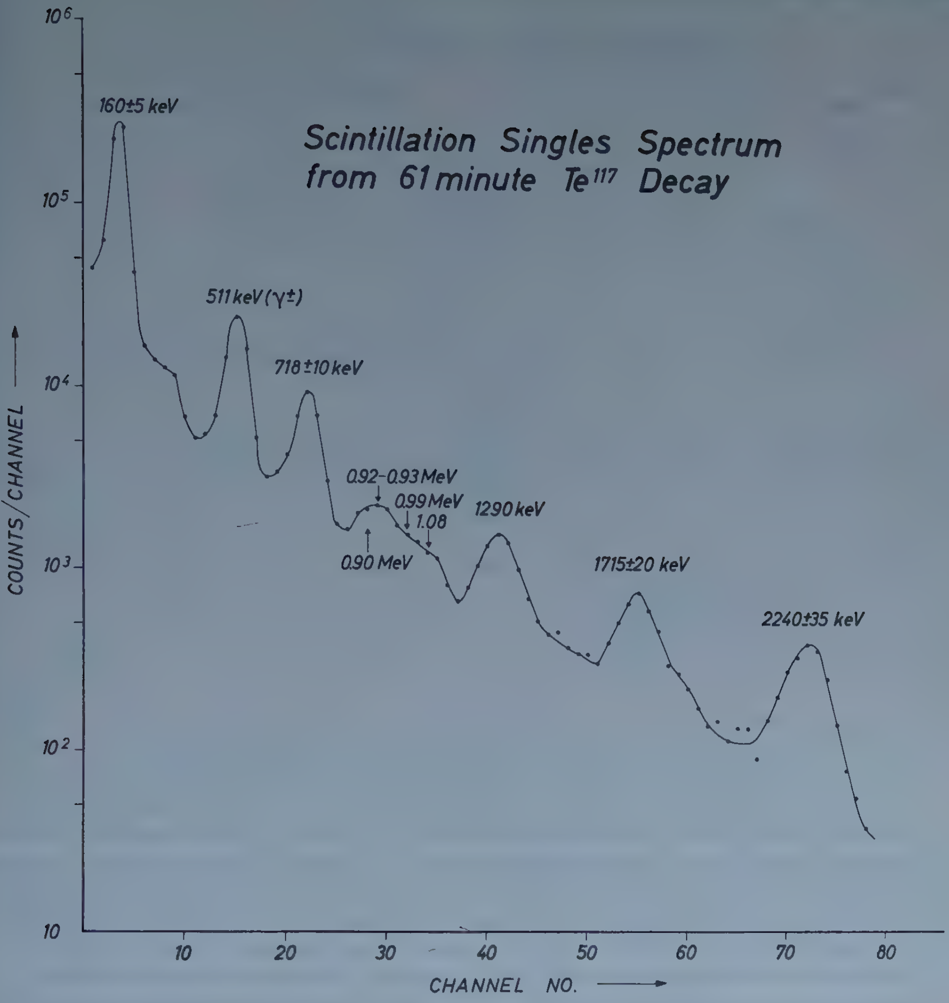

Fig. 3. Plot of the singles spectrum of mass-separated 61 min Te^{117}, with a source-to-crystal distance of 20 cm. Background is subtracted. The 0.72, 1.29, 1.71, and 2.24 MeV peaks and the 0.9–1.1 complex exhibited an initial half-life of about one hour (measured some 6.8 hours after mass separation). As the 61 min Te^{117} activity died out, the spectrum due to 2.5 hour Te^{116} became apparent, since the latter always was present in Te^{117} sources as a contaminant (e.g. 45 % of the total intensity in the 1.29 MeV peak at the time of measurement corresponding to about 0.7 % Te^{116} total contamination at the end of mass separation). (See discussion in text.) The 160 keV peak arises from the well-known transition in tin from the decay of 2.8 hour Sb^{117} daughter. The interpretation of the 0.9–1.1 MeV complex is discussed in the text. The 1.71 MeV peak arises in part from the escape of one annihilation quantum from pair production in the crystal due to the 2240 keV gamma.

region between 900 and 930 keV may contain more than one transition, but it was not possible to resolve it and so only one transition (920–930 keV) has been shown in the decay scheme. Also the region 990–1080 keV contains more than one transition only partially resolved in the coincidence spectra; for this reason, the 1080 keV transition is shown dashed in the decay scheme.

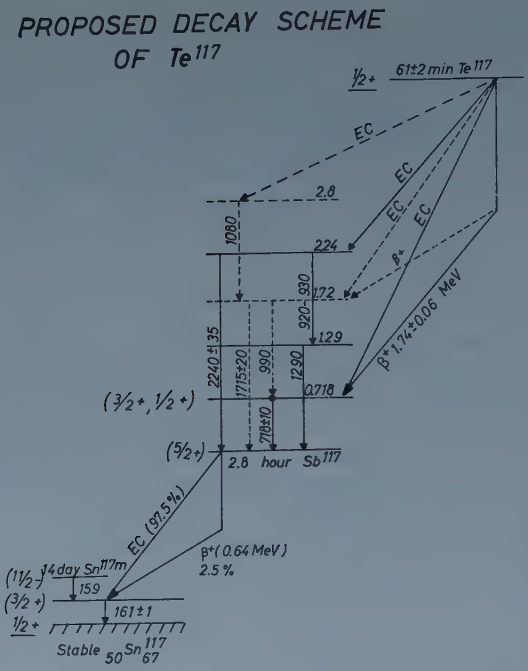

Fig. 4. Decay scheme proposed for Te^{117}. This scheme must be considered very tentative because the coincidence spectra could be studied in only one run. The spin of 1/2 for Te^{117} has been confirmed directly by the atomic beam magnetic resonance method.

The experimental result for the Sb^{117}/Te^{117} activity ratio, on the assumption that there is no direct ground-state feeding of Sb^{117}, is 8 ± 3 at the time of measurement. This ratio also can be computed from the time elapsed, 6.8 hours, between the measurement and an estimated "equivalent zero time" of mass separation; the computed value thus is 11 ± 2. The agreement within experimental error indicates that the assumption of no direct ground-state transition is tenable. However, the absence of the ground-state branch is not unambiguously proven because of the inaccuracy of the 161 keV relative intensity, although the resolution of the positron spectrum into two components about 1 MeV apart seems to support the level scheme (Fig. 4) in this respect.

From the relative intensities of the 161 keV transition in Sb^{117} and the 511 keV annihilation peak, together with the experimentally determined number of annihilation quanta detected per positron (2.86), and accepting a value 11 for the Sb^{117}/Te^{117} activity ratio at the time of measurement, an overall branching ratio $(EC/\beta^+) = 2.3$ is obtained, after allowing for 2.5% positron contribution from the Sb^{117} daughter [34]. If, according to the proposed decay scheme (Fig. 4), the 718 keV transition represents about 71% of the total decays, this ratio must arise primarily from decays leading to the 718 keV level.

Since the log ft value (5.1) for the 1.74 MeV positron branch clearly indicates an allowed transition with a theoretical K/β^+ ratio calculated to be 1.0 [26], L-capture contributing less than 15% [27], this is another case (cf. Te^{116}) of apparent disagree-

Table 2. Gamma–gamma coincidences from Te^{117}.

Gating energy (MeV)	Coincident gamma (MeV)
0.51 ($\gamma^{\pm}$)	0.72 (strong), 0.90
0.72	0.51 (strong), 0.99, 1.08
0.92 (?)[a]	0.51 (strong), 0.72[b], 0.90, 0.98, 1.29 (strong)
1.29	0.51 (strong), 0.92[a]
1.71	0.51, 1.08 (?)
⩾2	0.51 (?)[a]

[a] Due, at least in part, to Te^{116} contamination.
[b] Possibly due to the 0.9–1.1 MeV gamma complex.

ment between log *ft* and the EC/β^+ ratio as to the forbiddenness of the transition. However, rather large uncertainties enter the calculation of the experimental EC/β^+ value which may even bring the theoretical K/β^+ ratio within its limits of error.

Recently the spin of 61 min Te^{117} has been measured to be $\frac{1}{2}$ by the atomic beam magnetic resonance method by Dr. I. Lindgren *et al.*, Institute of Physics, Uppsala, in collaboration with one of the authors (R.W.F.). Thus it would seem on the basis of the log *ft* value for the 1.74 MeV positron branch that the 718 keV level in antimony has spin 3/2 + or 1/2 + (see Fig. 4). Insufficient information is at hand with which to suggest spin-parity assignments for the higher levels. Further discussion may be found in 4.2.

3.3. Te^{118}

Mass-separated sources exhibited a very straight gross beta decay in the beta proportional counter of half-life 6.00 ± 0.02 days, followed for about 8 half-lives.

The conversion electron spectrum was searched very closely in the beta spectrometer between 9 and 800 keV, and an extremely weak, unassigned, electron line of 19.6 keV energy decaying with a half-life of the order of 5 days was the only electron line observed, aside from the usual Auger complex. This line probably arises from *L* (23.8 keV) electrons from a trace of Te^{119} contamination. In particular, no lines were detected arising from the 40 and 255 keV transitions in tin which are fed only in the decay of 5.1 hour Sb^{118} [7, 35], confirming the absence of the latter in the 6 day Te^{118} decay. The intense positron spectrum was measured and gives an end-point of 2.59 ± 0.05 MeV. The *FK* plot indicates the presence of a weak inner group of end-point very roughly 1.2 MeV (allowed), although this may be inconclusive owing to poor statistics. In any case the inner branch seems to be much weaker in the *FK* plot than the inner branches found in Te^{116} and Te^{117}. The positrons probably arise from the 3.5 min Sb^{118} daughter, as shown in the decay scheme (Fig. 6).

From alpha spectroscopy an upper limit of $\alpha/\beta < 2 \times 10^{-8}$ could be set against conceivable alpha emission from Te^{118}.

From gamma singles scintillation spectra, which followed a 6 day half-life, the following gamma energies and relative intensities were measured: 511 ($\gamma^{\pm}$) (64), 830 ± 10, 1030 ± 15 (?), and 1220 ± 10 (1) keV. The 830 and 1030 keV peaks are very weak. No gammas having energies below 511 keV were found. Although the 1030 keV peak is reproducible, it is probably due, at least in part, to summing of annihilation quanta. The relative intensities of annihilation quanta and 1220

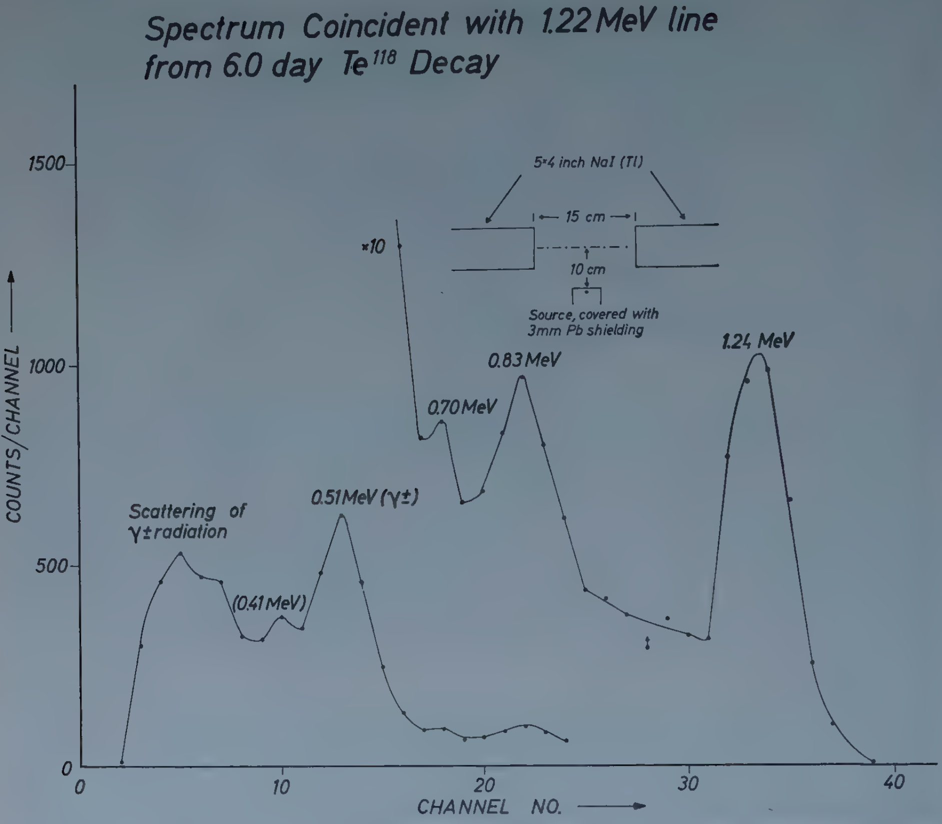

Fig. 5. Plot of a typical spectrum coincident with the 1.2 MeV line in Te^{118} decay The geometry is indicated in the figure. Two 5 × 4 inch NaI(Tl) crystals, 15 cm apart, were employed. The background and accidental coincidences were subtracted. The coincidence intensity was very weak so that counting times of 2 to 3 hours were necessary to obtain acceptable statistics, in contrast with the other tellurium isotopes studied in the present work where the coincidence intensity was much stronger. Two peaks are seen clearly, 0.83 and 1.24 MeV, to be coincident with the 1.2 MeV line, with a suggestion of a third peak at 0.41 MeV. In addition there is also a peak at 0.51 MeV due to positrons in coincidence with the 1.2 MeV line. Owing to scattering effects from the geometry used, the annihilation radiation exhibits a scattering distribution as shown on the figure and which is present with other positron emitters. The weak peak at 0.70 MeV is attributed to long-lived contamination. The Te^{118} coincidence spectra have been studied several times with sources from several different bombardments.

keV gammas correspond to a (β^+/1220 keV γ) ratio of 22, in agreement with the value of about 18.5 found by McGinnis and Kundu [35] who studied the 3.5 min Sb^{118} decay.

From this result, one calculates that the ratio of the number of positrons proceeding to the ground-state to the number going to the 1.22 MeV level in tin is approximately 24. Combining this with theoretical values for allowed transitions of $K/\beta^+ = 0.30$ and 2.1, respectively [26], one obtains a branching ratio of 90.4 % transitions ($K + \beta^+$) to the ground state and 9.6 % ($K + \beta^+$) to the 1.22 MeV level. Both transitions are allowed, since log $ft \sim 4.7$ for each.

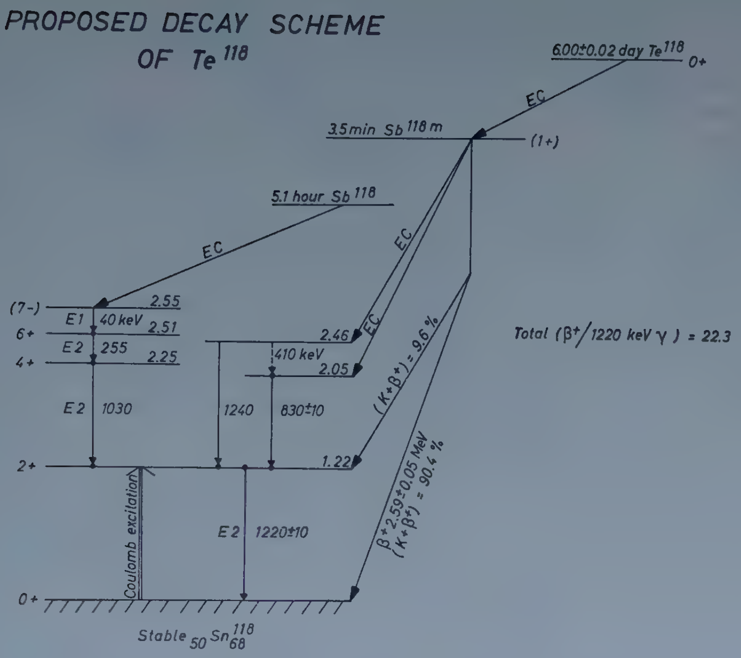

Fig. 6. Decay scheme proposed for Te^{118}. The very weak transition at 410 keV could be detected only in the spectrum coincident with the 1.2 MeV line, since it is otherwise masked by the very intense annihilation peak.

In coincidence spectra, shown in Fig. 5, the 511, 830, and additional gammas at about 410 and 1240 keV appeared in coincidence with the 1.2 MeV line. The relative intensities of these coincident peaks showed no change with successive coincidence spectra taken a few weeks apart. The 1030 keV gamma did not appear in the coincidence spectra. A tentative decay scheme is proposed in Fig. 6 to account for these data.

From the allowed beta decays to both the 0 + ground state and the well-established first excited 2 + level of Sn^{118}, 3.5 min Sb^{118} is a 1 + state. This assignment is further supported by the fact that no feeding of other levels in Sb^{118} from Te^{118} ground state is indicated; it is also in accord with the strong Nordheim rule [33]. The part of the decay scheme originating in 5.1 hour Sb^{118} is reproduced from [7] and included to demonstrate the absence of this activity in the Te^{118} decay. There is only weak evidence for the population of Sn^{118} levels above 1.22 MeV from 3.5 min Sb^{118}, and the log ft values for the two EC branches indicated cannot be calculated with sufficient accuracy to allow spin-parity suggestions to be made for the 2.05 and 2.46 MeV levels; see, however, the discussion in section 4.3.

3.4. Te^{119} *isomers*

Gross beta decay of mass-separated sources in the beta proportional counter gave two clear half-lives of 15.9 ± 0.3 hours and 4.5 ± 0.3 days, both followed for 5 half-lives, respectively. These correspond to the two known isomeric states of Te^{119}.

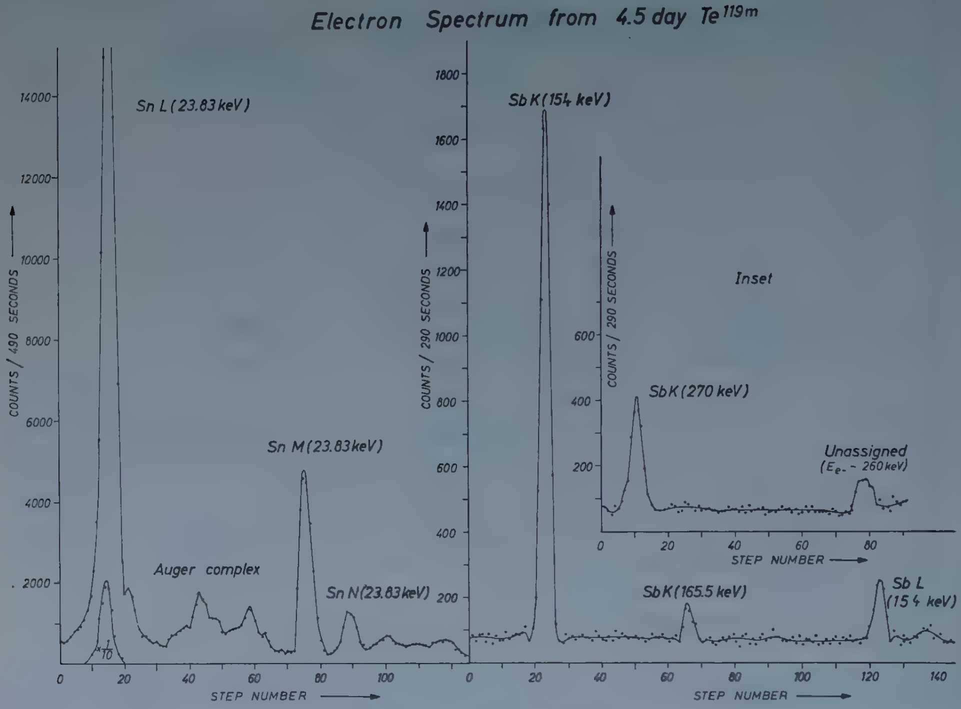

Fig. 7. Conversion electrons from 4.5 day Te^{119m} taken in the two-directional focusing beta spectrometer on sources sufficiently aged (about $2\frac{1}{4}$ days) that the 16 hour isomer essentially had decayed out. A thin-window (~ 50 $\mu g/cm^2$ VYNS) GM detector was used in the spectrometer. The time dependence of the L and M lines from the well-established 23.83 keV transition in tin, taken at approximately 0.5 % resolution, is shown in Fig. 8. The K and L lines from transitions in antimony at 153.7 and the K line from a transition at 165.5 keV were taken on a different source with 0.35 % resolution. The inset shows in addition a K line from a 270 keV transition, tentatively assigned in antimony, the L-line of which is not clearly seen because of masking by an unassigned line having roughly 260 keV electron energy. The half-life of the latter line was not established.

The beta spectrometer was used for close study of the electron spectrum between 9 and 440 keV. In addition, careful searches for positrons from the 4.5 day isomer were carried out in the spectrometer, but none were detected. The electron spectrum from 4.5 day sources, taken after the 16 hour isomer had decayed out, is shown in Fig. 7.

Conversion electrons corresponding to transitions in antimony at 153.7 ± 1.0, 165.5 ± 1.0, and 270 ± 2 keV were observed (Fig. 7) to follow a 4.5 day half-life. In particular, the K line of the 153.7 keV transition was followed in the beta spectrometer (resolution $\sim 1\%$) from a fresh mass-separated source of 16 hour + 4.5 day Te^{119}. There was no growth and no 16 hour component in the decay, a pure 4.5 day half-life being observed. Similarly, in the scintillation singles spectra (see below), no 16 hour component was observed in the decay of the 152, 270, 1214, and 2092 keV peaks. No other lines up to 440 keV were detected, except a weak, unidentified line of 260 keV electron energy, which conceivably could arise from conversion of a

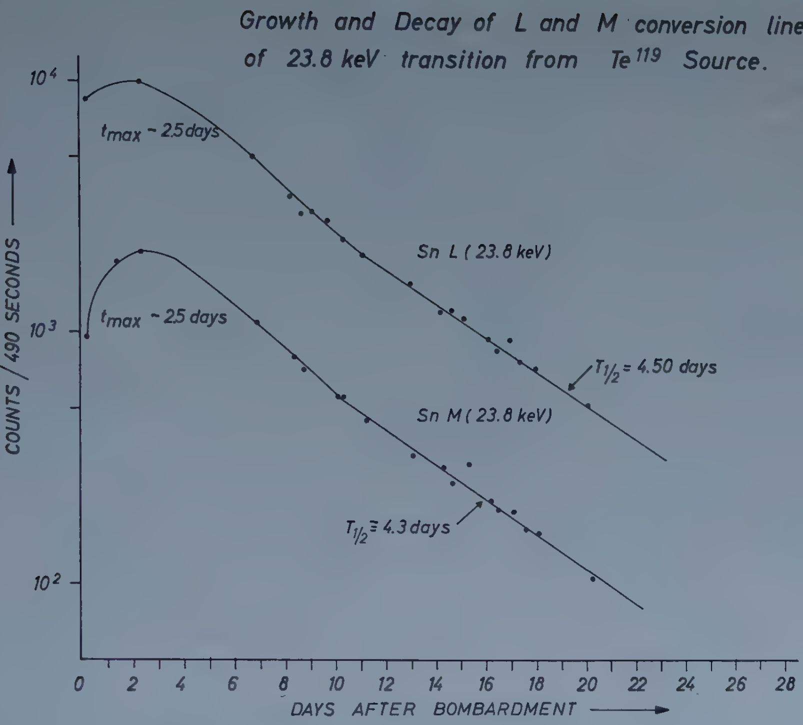

Fig. 8. Growth and decay of L and M lines corresponding to the well-established 23.83 keV transition in tin (see Fig. 7) from a source of 16 hour + 4.5 day Te^{119}. Following the point of maximum growth (2.5 days) which corresponds to that expected for a 4.5 day parent feeding a 38 hour daughter, the initial half-life is somewhat shorter than 4.5 days, indicating the contribution of 38 hour Sb^{119} from the 16 hour decay, but the final slope exhibits a 4.5 day half-life. The presence of Auger complex may account for the difference in the growth curves of the L and M lines.

290 keV transition, although the half-life was not followed. The K/L_I ratio of the 153.7 keV transition is 8.9 ± 0.3 (from Fig. 7), in agreement with $M1$ multipolarity for which the theoretical ratio interpolated from Rose's tables [25] is 8.97.

When a fresh source of 16 hour + 4.5 day Te^{119} was followed, the L and M lines from the well-established 23.83 keV transition in tin [36] followed the growth and decay curves shown in Fig. 8, in which the time of maximum growth (2.5 days) corresponds to that expected for a 4.5 day parent feeding a 38 hour daughter (Sb^{119}). Following this growth, the initial decay is somewhat faster than 4.5 days apparently because of that amount of the 38 hour daughter which arises from the independent decay of the 16 hour isomer, but the final slope, after decay of this extra accumulation of Sb^{119}, corresponds to a 4.5 day half-life.

The electron spectrum from a fresh source (16 hour + 4.5 day) is shown in Fig. 9, where the strongest peaks, other than the growing M and N conversion lines of the 23.83 keV transition in tin, followed a 16 hour decay, disappearing into an unresolved Auger complex, which finally followed a 4.5 day half-life. Relative intensities of the 16 hour lines correspond well to those expected for the principal K Auger transitions,

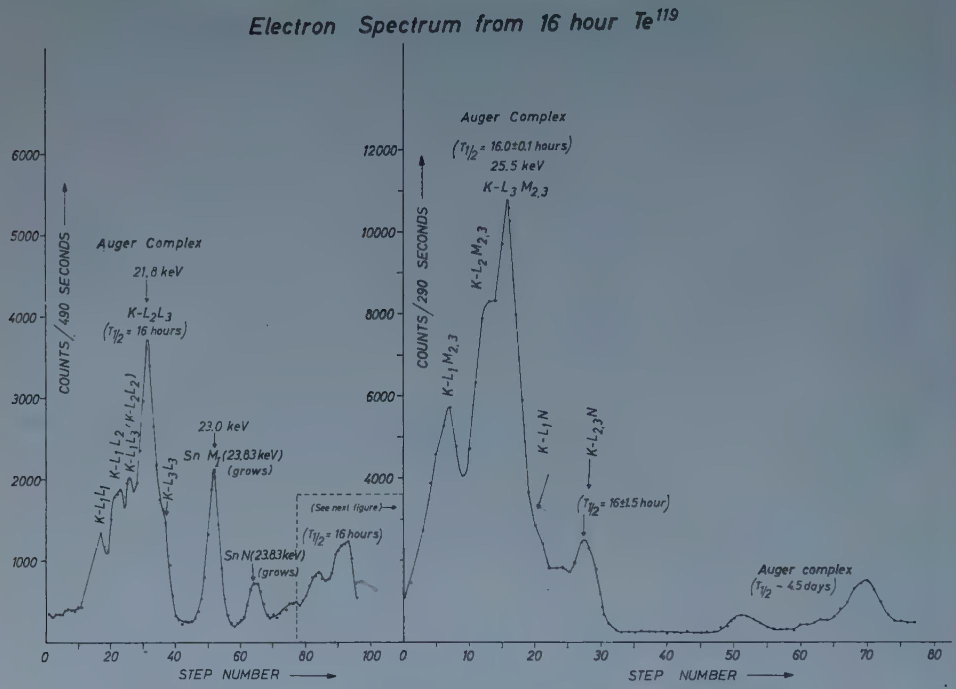

Fig. 9. Conversion electrons from a fresh source (~3 hours after bombardment) of mass-separated 16 hour + 4.5 day Te^{119}. A thin window (~50 μg/cm² VYNS) GM detector was used in the beta spectrometer, the resolution being set at approximately 0.5 % for the left-hand portion of the figure and 0.35 % for the right-hand section, which was taken with a much stronger source. The M and N lines of the well-known 23.83 keV transition in tin (Fig. 7) were observed to grow strongly (cf. Fig. 8). The strongest peaks in the Auger complex decaying with a 16 hour half-life correspond to the principal Auger transitions in antimony as listed in Table 3 of the text. The energies and relative intensities of these peaks agree better with their assignment as Auger transitions than as conversion lines of a possible 26 keV gamma transition in antimony.

Table 3. Principal Auger transitions expected in antimony (cf. Fig. 9).

Transition	Energy (keV) ($Z=51$) (calcd.)	Relative intensity		
		Calcd.[a] ($Z=47$)	Exptl.[a] ($Z=47$)	Exptl.[c] ($Z=56$)
$K-L_1L_1$	21.094	1.00	1.0	1.0
$K-L_1L_2$	21.412	1.82	1.3	1.4
$K-L_1L_3$	21.660	1.66	1.3	
$K-L_2L_3$	21.978	4.71	3.2	2.9
$K-L_3L_3$	22.226	1.90	1.8	1.5
$K-L_2M_{2,3}$	25.32	1.13	1.6	
$K-L_3M_{2,3}$	25.57	1.48	1.7	

[a] Theoretical results for $Z=47$ from [22].
[b] Experimental measurements for $Z=47$ from [23].
[c] Experimental measurements for $Z=56$ from [24].

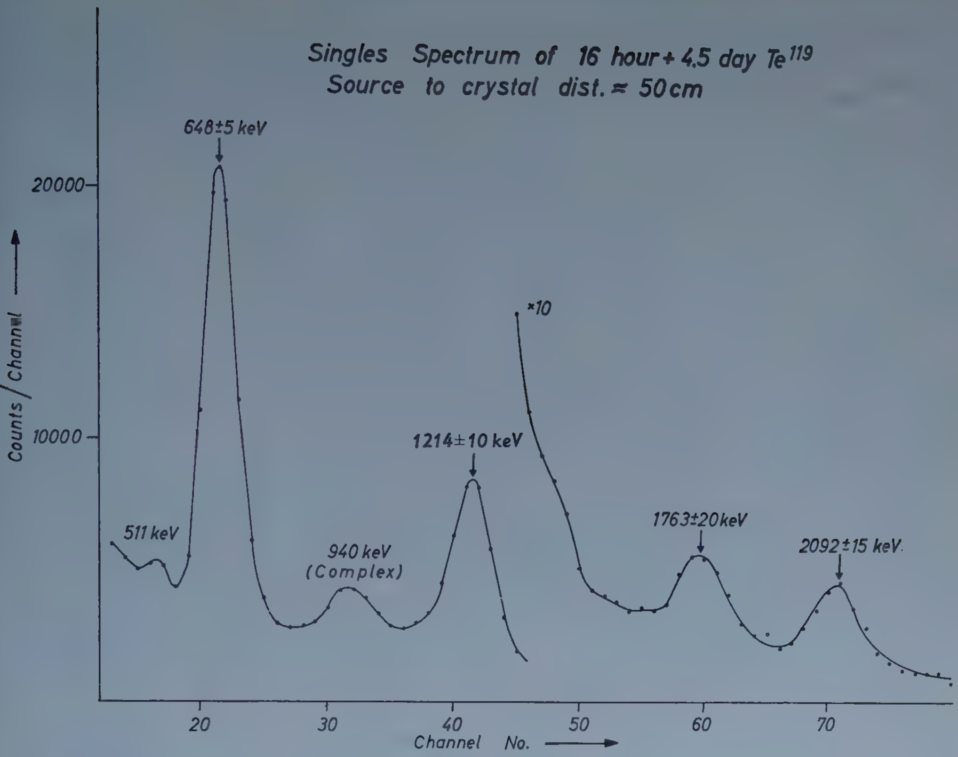

Fig. 10. Plot of the singles spectrum of 16 hour + 4.5 day Te^{119} isomers. The source-to-crystal distance was about 50 cm. The weak peak at 511 keV is due at least in part to annihilation radiation from Te^{118} impurity. The peak at 648 keV decayed with a pure 16 hour half-life. The 1763 keV peak also decayed with a 16 hour half-life, but with a weak residue decaying with a 4.5 day half-life. All other gammas, both in the singles and in coincidence spectra, decayed with a half-life of 4.5 days.

according to recent theoretical calculations of Asaad and Burhop [22] for $Z = 47$ and experimental values of Johnson and Foster [23] for $Z = 47$ and of Marguin and Moussa [24] for $Z = 56$. These literature data are summarized in Table 3 for comparison with Fig. 9.

From alpha spectroscopy [21], an upper limit of $\alpha/\beta < 4 \times 10^{-7}$ could be set against alpha emission above 1.9 MeV from Te^{119}, both isomers being taken together.

From scintillation singles spectra shown in Fig. 10 of fresh Te^{119} sources containing strong 16 hour activity, two gammas were measured and their relative intensities obtained: 648 ± 5 (95) and 1763 ± 20 (3) keV, the Cs^{137} gamma (661.4 keV) being used to calibrate the former energy. These two peaks exhibited a 16 hour half-life, but the 1763 keV peak also showed a very weak 4.5 day component. No gammas were found to be coincident with the 1763 keV transition, indicating that the latter proceeds directly to the ground state. The 648 keV gamma seems not to be in coincidence with any other hard gamma, but this is rendered somewhat inconclusive by the presence of higher energy gammas, although there is a clear absence of (2.1 MeV) (648 keV) coincidences.

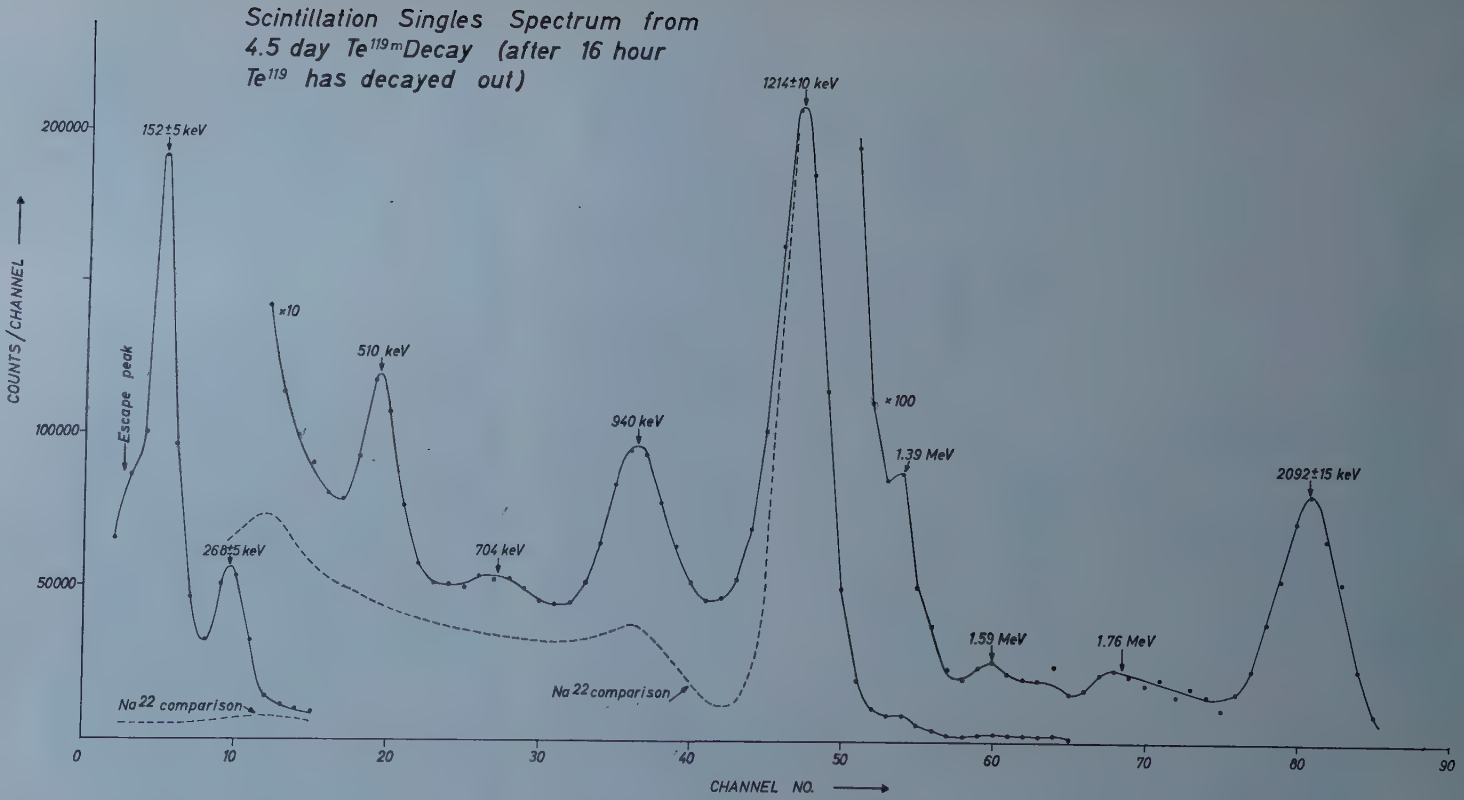

Fig. 11. Plot of a typical singles spectrum from 4.5 day $\mathrm{Te}^{119\,\mathrm{m}}$, taken at a time such that the 16 hour isomer had decayed out. This spectrum was studied a great many times with sources from six bombardments and the decay of all peaks was followed. An example of the method used to resolve the structure is indicated by the Na^{22} response function of the 1274 keV gamma fitted to the 1214 keV peak and shown dotted in the figure. From the singles spectrum the following gamma energies were determined: 152 ± 5 (which is more accurately determined from the electron spectrum to be 153.7 ± 1.0 keV), 268 ± 5 (270 ± 2 keV from the electron spectrum), 940 ± 20, 1214 ± 10, and 2092 ± 15 keV, using Na^{22}, Co^{60}, ThC'', $\mathrm{Te}^{123\,\mathrm{m}}$, and Hg^{203} as calibration sources. A very slight contribution at 1380 ± 20 keV is present from long-lived contamination. The peak at 511 keV is due mostly to annihilation radiation from Te^{118} present to a small extent in the Te^{119} sources as well as to annihilation resulting from pair production. There is an indication of some unresolved complexity in the region of 0.9–1.1 MeV which is resolved in the various coincidence spectra.

Table 4. Summary of gamma energies and relative intensities from 4.5 day Te^{119m}.

Energy (keV)	Relative gamma intensity (uncorrected for conv.)	Method of energy determination
153.7 ± 1.0^a	440 ± 150^a	Beta spectrometer (see Fig. 7)
165.5 ± 1.0^b	$\sim 30^b$	Beta spectrometer (Fig. 7)
270 ± 2^c	150 ± 50	Beta spectrometer (Fig. 7)
~ 400	Weak	Evident only in coin. spectra
918 ± 15	$\leqslant 100$	From coin. spectra
955 ± 15	~ 100	From coin. spectra
1000 ± 20	$\leqslant 100$	From coin. spectra
1130 ± 15	~ 50	From coin. spectra
1214 ± 10	320 ± 50	Calibration against Na^{22} and Co^{60} standards (Fig. 11)
1763 ± 20	1–3	From singles spectra of 16 hour isomer (Fig. 10), but also present weakly in 4.5 day decay (Fig. 11)
2092 ± 15	25 ± 5	Calibration against ThC″ and Na^{22} standards (Fig. 11)

[a] Also measured in the gamma singles spectrum (Fig. 11) as 152 ± 5 keV by comparison with a Te^{123m} standard (159.1 keV). The rel. intensity results from the 152 keV peak in the gamma singles spectrum when the calculated gamma intensity of the 165.5 keV transition (see *b*) is subtracted. The intensity when corrected for $M1$ conversion becomes 525 ± 150.

[b] Not resolved from the 152 keV peak in the gamma singles spectrum (Fig. 11). The rel. intensity is calculated from the ratio of its K conversion line and that of 153.7 keV, assuming $M1$ or $E2$ multipolarity for the 165.5 keV transition.

[c] Also measured in the singles spectrum (Fig. 11) as 268 ± 5 keV with an Hg^{203} standard (279.1 keV).

Table 5. Gamma–gamma coincidences observed from 4.5 day Te^{119m} (+ + indicates clear coincidence, + indicates less certain, − indicates clear absence of coincidence).

Coincident gamma (keV)	Gating energy (MeV)			
	0.15	0.27	1.2	2.0
154		+ +	+ +	
270	+ +	+	+	+ +
918			+ +	
955	+ +[a]	+ +	+	−
1000			+ +	
1130	−	+ +	−	−
1214	+ +			
2092		+ +	−	−

[a] 918, 955, and 1000 keV peaks were not resolved in spectrum coincident with the 0.15 MeV line.

Gamma energies and relative intensities (uncorrected for conversion) from singles spectra (Fig. 11) and coincidence spectra of well-aged 4.5 day Te^{119} sources, all 16 hour isomer having decayed out, are shown in Table 4. All principal gamma peaks were followed for several half-lives to establish their assignment to the 4.5 day decay.

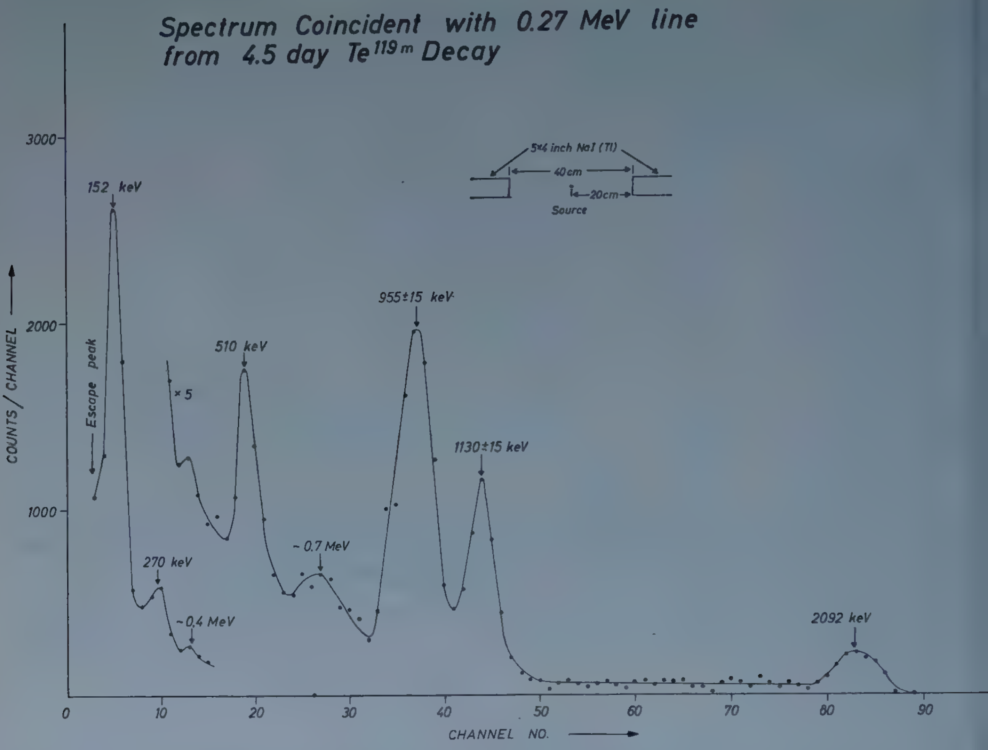

Fig. 12. Plot of a typical spectrum coincident with 0.27 MeV line from 4.5 day Te^{119m}, taken on sources from three bombardments at such times that all 16 hour isomer had died out. The geometry is shown in the figure. Energy calibration was obtained from that of the singles spectrum (Fig. 11) taken before and after the coincidence spectrum. Two new gammas are most clearly resolved in this spectrum; namely, those at 1130 ± 15 and 955 ± 15 keV. This spectrum was run at various delays, 0.3, 0.75, 0.90, 0.95, and 1.0 μsec, with no observed changes. The 0.51 MeV peak is interpreted to be due primarily to Te^{118} contamination (the Compton tail of the annihilation peak falling in the gate), with some additional contribution from annihilation of electron pairs produced by the 2092 keV gamma which is in coincidence with the 270 keV transition.

Besides the gammas listed in Table 4, peaks were observed at 704 and 1585 keV in the singles spectrum and in the spectrum coincident with a 0.51 MeV gating line. These are interpreted to arise from the escape of a single annihilation quantum from pair production in the crystal due to the 1214 and 2092 keV gammas, respectively. The 510 keV peak (Fig. 11) arises partly from annihilation radiation due to the presence of a small amount of Te^{118} contamination and partly from annihilation of pairs produced from the Te^{119} source by the 1214 and 2092 keV gammas.

In Table 5 the qualitative results of coincidences with lines at 0.15, 0.27, 1.2, and 2.0 MeV are summarized for measurements on the 4.5 day state. In interpreting the results, however, it should be noted that in certain cases it has not been possible to allow for tails of other gammas falling in the gating channel, and this may cause some discrepancies in the qualitative strengths of the coincidence intensities in the case of weak coincidences. All coincidence spectra were followed for decay to confirm the 4.5 day half-life for each coincident peak.

Table 6. Gamma energies and relative intensities in spectrum coincident with 0.27 MeV line from 4.5 day Te^{119m} (Fig. 12).

Energy (keV)	Relative intensity
152	91 ± 35
270	10 ± 5
955 ± 15	63 ± 20
1130 ± 15	29 ± 10
2092	14 ± 5

Relative intensities of gammas in coincidence with a 0.27 MeV gating energy are summarized in Table 6 from data shown in Fig. 12. The presence of the relatively strong 270 keV coincidence is somewhat surprising. This may be explained as being due to Compton and back-scattering tails from higher energy gammas falling in the gating channel and would thus indicate the possibility that the 152 keV coincidence peak receives a similar contribution. A correction for this effect may bring its intensity into better agreement with that of the 955 keV coincidence peak as needed for the proposed decay scheme (Fig. 13). Another possible explanation would be the existence of a second transition of about 270 keV energy. In this connection it may be recalled that an unassigned line in the Te^{119m} electron spectrum (Fig. 7) would correspond to a transition energy of 290 keV if interpreted as a K conversion line in Sb. That this line belongs to the Te^{119m} decay, however, is not established experimentally.

The spins of the 16 hour and the 4.5 day isomers have been measured by the atomic beam magnetic resonance method to be 1/2 and 11/2, respectively (Dr. I. Lindgren *et al.*, Institute of Physics, Uppsala, in collaboration with one of the authors (R.W.F.)).

A tentative decay scheme for the Te^{119} isomers is proposed in Fig. 13, in which the weak 918 keV transition has not been placed. It is fairly clear that the 4.5 day (11/2 −) isomer is the upper state since the ground-state Q-value for the 16 hour decay is $\leqslant 2.3$ MeV, based on the positron measurement of Kocher, Mitchell, Creager and Nainan [10], which is exceeded by that of the 4.5 day decay on any of the possible arrangements of the gamma transitions in Sb. From level systematics (cf. 4.1), the energy difference 11/2 − to 1/2 + in Te^{119} would be about 320 keV, leading to a Q-value for the 4.5 day state of 2.62 MeV, which should be considered an puper limit.

The Te^{119m} decay scheme (Fig. 13) seems to be internally consistent and to account for the main experimental findings of the present work. In particular it should be pointed out that the absence of beta branches to Sb levels lower than 1.37 MeV explains the failure to find positrons in the decay. However, the scheme is not in accord with the triple coincidence (1.22, 0.930, 0.153) reported by Kocher *et al.* [10] if their 0.930 is identified with our 955 keV transition, which is predominant in the 0.9–1.1 MeV complex. The indication (Table 5) of a coincidence (1.2, 0.918) may resolve this discrepancy, suggesting that the 918 keV gamma feeds the 1.37 MeV level. Another difficulty concerns the depopulation of this level. The log ft value (~ 6.3) obtained for its EC feeding on the basis of the proposed decay scheme and the estimated disintegration energy is consistent with a first-forbidden transition, implying an 11/2 + or 9/2 + assignment for the 1.37 MeV level. From the $M1$

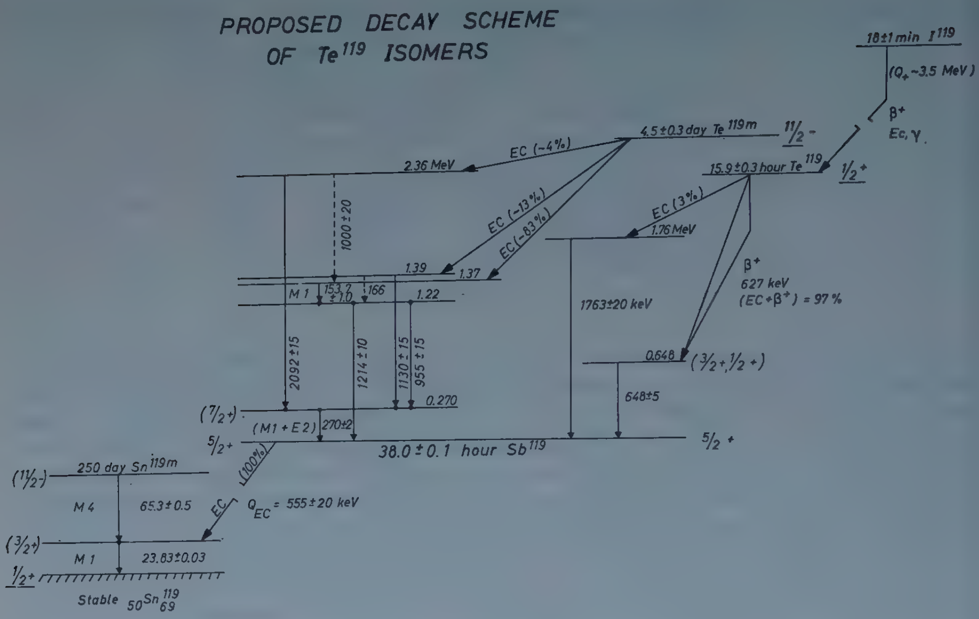

Fig. 13. Decay scheme proposed for Te^{119} isomers. The 955 and 1214 keV transitions are placed on the basis of coincidence information and energy sums, whereas the positions of the 153.7 and 1130 keV transitions depend chiefly on coincidences. The weak 918 keV transition has not been placed, since present information is inadequate to suggest its position. Incoming and outgoing intensities for the various levels seem to be consistent within error limits. No beta feeding is required for the 0.270 or 1.22 MeV levels, since they are connected internally, and the 270 and 1214 keV transitions both go to the ground state. If the lowest level fed directly from 4.5 day Te^{119m} is at 1.37 MeV, this explains the absence of positrons in the 4.5 day decay, assuming $Q+\approx 2.6$ MeV.

character of the 154 keV transition, the 1.22 MeV level would then be 9/2 + or 7/2 +, respectively. Absence of appreciable *EC* branching to this level makes 7/2 + the more probable, and hence 9/2 + is suggested for the 1.37 MeV level. This, however, is incompatible with the fact that it feeds the 1.22 MeV level by the strongest transition found in the decay and yet does not to any detectable degree feed the strongly energy-favored 0.270 MeV level which for good reasons (cf. 4.2) is likewise assigned 7/2 +. Consequently, it seems that the 1.37 and 1.22 MeV levels after all should be 11/2 + and 9/2 +, respectively. In addition, it is necessary to assume widely different excitational character of the levels concerned, thus putting strong selection rules into operation both for the beta and gamma decay. In view of these difficulties we have refrained from suggesting spin and parity for the higher levels. It may be remarked that log *ft* for the *EC* branch leading to the 2.36 MeV level, as calculated on the above assumption about the *Q*-value, becomes roughly the same as for the one feeding the 1.37 MeV level.

The decay of the 16 hour Te^{119} ground state seems much more clear-cut. On the reasonable assumption that the 627 keV positron branch found by Kocher *et al.* [10] populates the same Sb level (0.648 MeV) as the predominant *EC* branch, its log *ft* is calculated to be 5.6, using the measured relative intensity of the 648 keV gamma and the theoretical K/β^+ ratio. Thus $3/2+$ or $1/2+$ is suggested for the 0.648 MeV level. As discussed in section 4.2, the former assignment is probably to be preferred. For the 1.76 MeV level the log ft value (6.1) of the EC branch does not give an unambiguous indication of the spin-parity assignment. It may result from 1-forbiddenness, although the 627 keV positron branch, which on the assumption that the 0.648 MeV level is $(d)3/2+$ also would be 1-forbidden for decay from $(s)1/2+$ Te^{119}, shows a normal allowed log ft value (5.6). That an 1115 keV stop-over transition was not detected in the present experiments is not conclusive because of the presence of the fairly strong 4.5 day 1130 keV gamma.

3.5. *Other activities observed*

In preliminary experiments in which antimony activities were mass-separated using metallic antimony radiochemically separated from a bombardment of tin metal with 110 MeV protons, gross beta decay gave half-lives of 36 ± 3 min for Sb^{115} and 64 ± 4 min for Sb^{116m}. The former confirms the recent Russian results [6].

Preliminary studies of short-lived tellurium activities, radiochemically separated by the 30 sec procedure described above from NH_4I irradiated for 10 min with 145–180 MeV protons, indicated the presence of a new activity of tellurium with half-life about 5–6 min, from gross decay in the beta proportional counter and in the alpha ionization chamber. The new activity may be due to Te^{115} or Te^{114}.

4. Discussion

4.1. *Isomerism in odd-*A *tellurium isotopes*

A smooth continuation toward lower mass numbers of the systematics of low-lying levels in odd-A tellurium isotopes [37] predicts an $M4$ transition of about 80 keV in Te^{119}. If the matrix element is the same as for the higher mass tellurium isomeric transitions [38], the expected internal branch from 4.5 day Te^{119m} is calculated to be about 3 %. Because of the high conversion of an 80 keV $M4$ transition, however, its K conversion line would be more than 100 times stronger than K 153.7 keV, which is the most prominent line observed in the electron spectrum (Fig. 7). In the same way, the possibility of an $M4$ transition of any other energy within the region studied is ruled out, and we conclude that the level spacing $11/2-$ to $3/2+$ in Te^{119} is less than about 15 keV and very probably negative. This indicates a marked discontinuity in the level systematics, for which there seems to be no obvious explanation.

There is a second possibility for internal decay from the $11/2-$ state, namely by an $E5$ transition to the $1/2+$ ground state. Its energy, as indicated by a smooth systematics, would be about 320 keV, which implies a very weak branch assuming a reasonable value of the matrix element. As an $E5$ transition of this energy has relatively low conversion, moreover, such a transition easily could have escaped detection in the present electron spectra.

The failure to find a metastable state of half-life >15 minutes in Te^{117} can be explained in several ways, most simply perhaps by assuming a level crossing in tellurium in which the $5/2+$ level passes below the $11/2-$ and gives rise to a rather short-lived $E3$ transition. This would be in agreement with the trend of decreasing

5/2 + to 1/2 + level spacings toward lower A, as indicated both by experimental and theoretical results for Te^{125} and Te^{123} [9]. On the other hand, the increase in the beta disintegration energy in going from Te^{119} to Te^{117} may be sufficient to cause the decrease in half-life indicated; it may be noted that the partial EC half-life of Te^{121m} is as high as 3080 days which makes the EC half-life ratio Te^{121m}/Te^{119m} about 700. In addition, the high disintegration energy of Te^{117} may make an 11/2 − proton level in antimony available for allowed decay.

4.2. *Levels in odd-A antimony isotopes*

The ground states of Sb^{123} and the higher odd-A Sb isotopes are known to be 7/2 +, whereas Sb^{121}, Sb^{119}, and, tentatively, Sb^{117} are assigned 5/2 +. In these latter isotopes, 7/2 + is likely to appear as a low-lying excited state and in fact has been identified by Gupta [39] with a 70 keV level in Sb^{121} which is fed in the decay of both the 17 day Te^{121} ground state and the 154 day metastable state. No corresponding low-energy transition is found in the decay of 16 hour Te^{119}, although, according to the proposed decay scheme (Fig. 13), 4.5 day Te^{119m} feeds a level at 270 keV in Sb^{119} which can be given a 7/2 + assignment. The de-excitation, $g_{7/2}$ to $d_{5/2}$ in single particle notation, would then be 1-forbidden for an $M1$ transition. This agrees with the mixed $M1+E2$ character of the 270 keV gamma indicated by the conversion data of Kocher *et al.* [10].

The 270 keV Sb^{119} level as well as the 70 keV Sb^{121} level is seen to fit rather nicely into the 7/2 + to 5/2 + energy systematics shown in Fig. 14. The identity of the levels cannot be considered satisfactorily proven, however. In particular, the apparent difference between the ground-state decays of Te^{121} and Te^{119} needs some discussion. The levels at 576 and 648 keV predominantly fed in Sb^{121} and Sb^{119}, respectively, are probably analogous (see below). The former, however, decays partly by a ground-state transition and partly by a 506 keV transition to the 70 keV level, the intensity ratio being 100/20 [39], whereas in the Sb^{119} case neither a 378 keV transition between the 0.648 and 0.270 MeV levels nor a 16 hour component in the 270 keV peak could be established. Assuming equal reduced transition probabilities for corresponding transitions in the two cases, one calculates an expected intensity ratio 648 keV/378 keV ~ 100/3. This means that the hypothetical 378 keV peak in the 16 hour scintillation spectrum would be of the same relative intensity as the 1763 keV peak but would almost certainly have escaped detection owing to the presence of Compton distributions from the 648 keV gamma and the strong 4.5 day gammas. Similarly, it would require great accuracy to reveal a 16 hour component of such low intensity in the 4.5 day 270 keV peak.

From the presence of the 506 keV stop-over transition to the 7/2 + state in Sb^{121}, the spin-parity assignment 3/2 + is preferred for the 576 keV level, excluding 1/2 + which is otherwise an alternative because of the allowed feeding from 1/2 + Te^{121} [39]. According to these considerations, the same argument should apply to the assignment of the 648 keV level in Sb^{119} and also of the 718 keV level in Sb^{117}. The systematic behaviour of the energies of these levels (Fig. 14) may be taken as support for their identification as corresponding excitations of these three nuclei.

In the case of Sb^{117}, the position of the first excited 7/2 + level is not known, owing to the failure to find an 11/2 − metastable state in Te^{117}. It is interesting to note that the levels in Sb^{117} at 1.29 and 2.24 MeV are similar both in energy and in

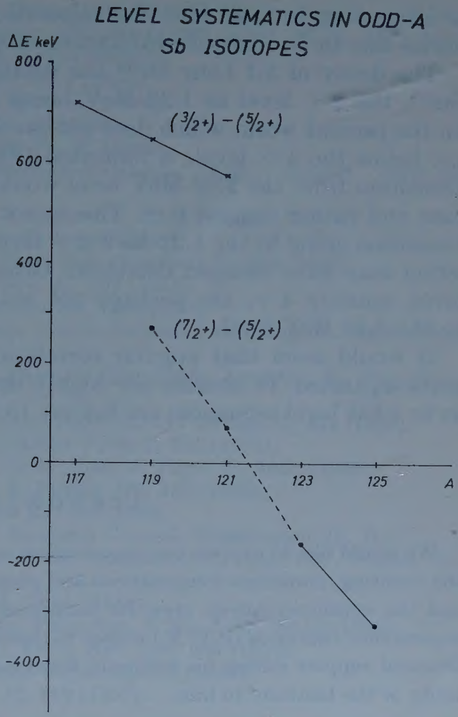

Fig. 14. Systematics of level spacings in odd-A Sb isotopes interpreted to be (g) 7/2 + to (d) 5/2 + and (d) 3/2 + to (d) 5/2 +.

mode of depopulation to the lowest excited states in Sn^{116} fed from 15.5 min Sb^{116}, which might be a reflection of the fact that Sb^{117} comprises an odd proton coupled to an Sn^{116} ($Z = 50$) core.

4.3. *Levels in even-*A *Sn isotopes*

In Sn^{116} excited 2 + levels at 1.30 and 2.12 MeV have been established by several studies of 54 min In^{116m}, most recently by Girgis and Van Lieshout [31]. The first excited state is known to be fed also from 15.5 min Sb^{116}, whereas the next higher level populated in this decay has been placed approximately 100 keV above the 2.12 MeV 2 + level. The present work confirms this difference in the level sequences and gives, moreover, good evidence that the 2.23 MeV level also is 2 + (cf. 3.1). The 930 keV transition depopulating this level is not found in the In^{116m} decay ($< 1\,\%$) [31]; similarly, we can set an upper limit of a few percent against feeding of the 2.12 MeV level from the Sb side. Thus the two close-lying 2 + levels must be of quite different character. Their properties can be compared by means of the excitation energies relative to that of the first excited state and the intensity ratios of stop-over and cross-over transition as follows:

$$\text{2.12 MeV level: } E_2/E_1 = 1.64,\ I(\text{stop-over})/I(\text{cross-over}) = 0.88,$$
$$\text{2.23 MeV level: } E_2/E_1 = 1.73,\ I(\text{stop-over})/I(\text{cross-over}) = 1.78.$$

It seems difficult on the basis of these data to decide which of them should be interpreted as a "two-phonon" vibrational level. As to the character of the other, it is interesting to note that the calculations by Kisslinger and Sorensen [8] predict states due to two-particle excitation close to 2 MeV in Sn^{116}.

The decay of 5.1 hour Sb^{118} has established the level sequence 0 +, 2 +, 4 + in Sn^{118}, the 2 + level at 1.22 MeV being fed also from the 3.5 min isomer of Sb^{118}. In the present work, which does not involve 5.1 hour Sb^{118}, a new level at 2.05 MeV, i.e. below the 4 + level, is indicated (Fig. 6). The apparent lack of a ground-state transition from the 2.05 MeV level would seem to exclude a 2 + assignment in this case and rather suggest 0 +. This is not conclusive, however, because the 830 keV transition going to the 1.22 MeV 2 + level is weak and a still weaker cross-over transition may have escaped detection. Other spin-parity assignments of the 2.05 MeV level, notably 4 +, are perhaps not entirely excluded. Similar speculations apply to the 2.46 MeV level.

It would seem that angular correlation measurements on Sn^{116} and Sn^{118} using mass-separated Te sources are highly desirable in order to resolve the ambiguities as to what level sequences are fed via 15.5 min Sb^{116} and 3.5 min Sb^{118}, respectively.

ACKNOWLEDGEMENTS

We would like to express our appreciation to Mrs. Anita Åström for assistance with some of the counting, numerous computations and preparation of figures. We thank Ing. Åke Svanheden and the synchrocyclotron crew for numerous bombardments, and Ing. Rolf Ringh for mass separations. One of us (R.W.F.) wishes to thank the Swedish Atomic Research Council for partial financial support during his tenure in Sweden and Prof. The Svedberg for extending the hospitality of the Institute to him.

The Gustaf Werner Institute for Nuclear Chemistry, University of Uppsala, and Institute of Physics, University of Helsinki, November 1960.

REFERENCES

1. LINDNER, M., and PERLMAN, I., Phys. Rev. *73*, 1124 (1948); *78*, 499 (1950).
2. DROPESKY, B., Thesis, Univ. of Rochester, N.Y. (1953) (unpublished).
3. FINK, R. W., Thesis, Univ. of Rochester N.Y. (1953) and FINK R. W., and WIIG, E. O., Phys. Rev. *94*, 1357 (1954).
4. KUZNETSOVA, M. YA., MEKHEDOV, V. N., RYBAKOV, V. N., and KHALKIN, V. A., Atomnaya Energiya *4*, 583 (1958) (in Russian).
5. RHODES, A., Univ. of California report UCRL-3879 (1957) (unpublished).
6. SELINOV, I. P., GRITS, IU. A., KHULELIDZE, D. E., BARONI, E. E., BLIODZE, IU. P., D'OMIN, A. G., and KUSCHAKEVICH, IU. P., Atomnaya Energiya *5*, 660 (1958) (translation in Reactor Science *11*, 176 (1960)).
7. SKYTTE JENSEN, B., NIELSEN, O. B., and SKILBREID, O., Nuclear Phys. *19*, 654 (1960).
8. KISSLINGER, L. S., and SORENSEN, R. A., Mat. Fys. Medd. Dan. Vid. Selsk. *32*, No. 9 (1960).
9. GLENDENNING, N. K., Phys. Rev. *119*, 213 (1960).
10. KOCHER. C. W., MITCHELL, A. C. G., CREAGER, C. B., and NAINAN, T. D., Phys. Rev. *120*, 1348 (1960); private communication from Prof. A. C. G. MITCHELL (1960).
11. ANDERSSON, G., Arkiv Fysik *12*, 331 (1957). BURGMAN, J. O., and ANDERSSON, G., Nuclear Instr. *3*, 33 (1958).
12. JUNG, B., Nuclear Phys. *10*, 440 (1959). JUNG, B., and ANDERSSON, G., Nuclear Phys. *15*, 108 (1960).
13. LEE, D., and COOK, G. B., AERE-C/R-430 (1950). JOLIOT, F., J. chim. phys. *27*, 119 (1930). NORWITZ, G., Anal. Chim. Acta *5*, 109 (1951). ALVÄGER, T., and OELSNER, G., Arkiv Fysik *12*, 319 (1957). OELSNER, G., Priv. communication (1959).

14. SMITH, G. W., and REYNOLDS, S. A., Anal. Chim. Acta *12*, 151 (1955). FREILING, E. C., PASCUAL, J., and DELUCCHI, A. A., USNRDL-TR-231 (1958). Šušić, M. V., Bull. Inst. Nuclear Sci. "Boris Kidrich" (Belgrade), *7*, 39 (1957).
15. ARBMAN, E., and SVARTHOLM, N., Arkiv Fysik *10*, 1 (1956).
16. ARBMAN, E., and JUNG, B., Nuclear Instr. & Meth. (in press).
17. PATE, B. D., and YAFFE, L., Can. J. Chem. *33*, 15 (1955).
18. KANTELE, J., Ann. Acad. Sci. Fennicae, Ser. A, VI, 37 (1959).
19. GOVE, H. E., and RUTLEDGE, A. R., Atomic Energy of Canada, Ltd., Report CRP-755 (1958).
20. EVANS, R. D., The Atomic Nucleus, p. 717. McGraw-Hill Book Co. Inc., 1955.
21. KARRAS, M., NURMIA, M., and ANDERSSON, G. To be published.
22. ASAAD, W. N., and BURHOP, E. H. S., Proc. Phys. Soc. (London) *71*, 369 (1958).
23. JOHNSON, F. A., and FOSTER, J. S., Can. J. Phys. *31*, 469 (1953).
24. MARGUIN, P., and MOUSSA, M. A., J. phys. radium *21*, 149 (1960).
25. ROSE, M. E., Internal Conversion Coefficients. North-Holland Publishing Co., Amsterdam 1958.
26. FEENBERG, E., and TRIGG, G., Revs. Modern Phys. *22*, 399 (1950). ZWEIFEL, P., Phys. Rev. *107*, 392L (1957); *96*, 1572 (1954).
27. ROBINSON, B. L., and FINK, R. W., Revs. Modern Phys. *32*, 117 (1960); *27*, 424 (1955).
28. SLÄTIS, H., DUTOIT, S. J., and SIEGBAHN, K., Arkiv Fysik *2*, 321 (1951).
29. SINGH, P. P., DOSSO, H. W., and GRIFFITHS, G. M., Can. J. Phys. *37*, 1055 (1959).
30. STÄHELIN, P., MAEDER, D., and POCHON, M., Z. Physik *140*, 498 (1955).
31. GIRGIS, R. K., and VAN LIESHOUT, R., Physica *25*, 590 (1959).
32. Nuclear Data Sheets, No. 58–5–66, National Research Council, Washington 25, D.C.
33. WAY, K., KUNDU, D. N., MCGINNIS, C. L., and VAN LIESHOUT, R., Ann. Rev. Nuclear Sci. *6*, 129 (1956).
34. MCGINNIS, C. L., Phys. Rev. *97*, 93 (1955); *94*, 780A (1954).
35. MCGINNIS, C. L., and KUNDU, D. N., Bull. Am. Phys. Soc. *3*, No. 1, 62, VA10 (1958).
36. OLSEN, J. L., MANN, L. G., and LINDNER, M., Phys. Rev. *106*, 985 (1957).
37. GOLDHABER, M., and HILL, R. D., Revs. Modern Phys. *24*, 179 (1952).
38. ALVÄGER, T., and OELSNER, G., Arkiv Fysik *12*, 319 (1957).
39. GUPTA, R. K., Nuovo cimento *18*, 665 (1960).

Tryckt den 13 april 1961

Uppsala 1961. Almqvist & Wiksells Boktryckeri AB